

NO_x EMISSION IN IRON AND STEEL PRODUCTION: A REVIEW OF CONTROL MEASURES FOR SAFE AND ECO-FRIENDLY ENVIRONMENT

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Abstract

Iron and steel manufacturing involved preparation of raw materials through processes such as sintering, pelletizing and coke making. During these processes, pollutants such as Sulphur (iv) oxides (SO₂) Carbon II oxides (CO), Nitrogen oxides (NO_x), Volatile organic compounds (VOC) and Particulate matter (PM) etc. are emitted. The present work is aimed at describing some mitigation technologies of controlling NO_x emissions in iron and steel production. The processes involved in the production of iron and steel using Blast Furnace (BF) and Basic Oxygen Furnace (BOF) has been described. The mitigation technologies of controlling NO_x emissions were analyzed and discussed with environmental impacts based on the economical and technical factors. In this work, the data presented is based on existing reviews. The combination of low NO_x burner (LNB) and Selective catalytic reduction (SCR) is capable of reducing NO_x emission for up to 90% and above. Emissions of other pollutants into the atmosphere as a result of ammonia slip, formation of acids and other gases are harmful to the environment and causes damage to the SCR systems. Installation and operation cost are the major impacts of the SCR technology in the process of iron and steel production.

Keywords: Emissions, Iron and steel, Nitrogen oxides, Selective catalytic reduction.

1. Introduction

Iron and steel are essential commodities that are widely used as industrial raw material, constructional material in high way networks and bridges, defensive hardware in villages and cities etc. Among the most important industries worldwide is Iron and steel industry. The annual demand for world steel grow approximately from 1.410 million tons of crude steel in 2010 (USGS, 2012) to 2200 million tons in 2050 (Bellevrat and Menanteau 2008) accounting for the emissions of pollutants in the atmosphere. The processes involved in raw material preparation for the manufacture of iron and steel are sintering, pelletizing and coking. This chemically reduces iron to molten iron in the BF or BOF and direct or smelting reductions to produce steel (EIPPCB, 2010). Steel can also be manufactured by direct scrap in an electric arc furnace (EAF). The present work study the processes involved in the production of iron and steel using the BF/BOF and EAF. The study investigates on emissions produced by the processes, how they are produced and the impacts of the emissions on the environment. The objective is to analyze control measures of NO_x emission in iron and steel production and similar processes based on data of existing reviews.

1.1 Iron and Steel Making Processes

The basic material required in making iron and steel consist of iron ore which is the main constituent element in the process. Coke is used to convert iron ore into iron. It is the residue obtained in heating of the pulverized coal gradually to high temperature of (900-1100°C) in the absence of air for approximately 18 hours (U.S. EPA, 2012). The process is enhanced by limestone combined with gangue in the coke making. Fluid slag can readily be separated from the hot iron are charged into the blast furnace to produce iron. In steel making, BF or BOF and EAF systems are the

most commonly used nowadays (Li *et al.*, 2017). An analysis to comparatively investigate energy efficiency, material efficiency and emission intensity at enterprise level. The emission intensity of dust, CO₂, SO₂, NO₂ and CO of the BF/BOF route is 7.7, 2.6, 92.6, 33.5 and 12.0 times greater than those of the EAF route respectively.

In 2010, the production of steel using BF/BOF and EAF accounted for approximately 65% and 30% worldwide (World steel, 2011). BF/BOF is usually applied for commercial quantity due to its large output steel making facilities. EAF is flexible in the use of raw material, low construction cost, and improved process control (U.S. EPA, 2012). The operational capability is below full capacity and has the ability to produce steel over a wide composition range.

Iron and steel production usually involved sintering, pelletizing and coke making processes. In sintering, raw materials and scrap are blended and combusted. At beginning of the sinter, a burner hood ignites the coke in the mixture providing sufficient heat from (1300-1480)⁰C to cause agglomeration and surface melting of the mixture, converting the fines into coarse lumps (Ooi, *et al.*, 2011). The purpose of sintering is to form dust, ore and coke fines into particles of suitable size, strength and porosity. Its disadvantage is that it requires large capital investment for air pollution control measures (APP, 2010). Iron ores are grounded into suitable particle size allowing the concentrated iron oxides content to be agglomerated and charged into the furnace. Such particles are formed as green ball called pellet. This involved the use of binders holding together the iron oxide grains rolled into ball by pelletizing disc that produce high strength pellets. The pellets are then hardened after drying off and finally heated to temperature of about 1300⁰C (Eisele and Kawatra, 2003). Before final hardening, a binder is needed to stick the grain particle together as soon as the pellet gets dried.

Coke is an essential raw material for the production of iron and steel. About 65% of the world steel production use coke to produce molten iron in the blast furnace. Coke is obtained by cooking of coal to drive off the impurities that will hinder the process in iron production. During the heating process, the physical form of the coke swells up and is important in maintaining the good condition in the process (Michael and Manfred 2012). Coke is added in the blast furnace to maintain the gas flow throughout the reacting material to reduce iron (U.S. EPA, 2012).

1.2 Iron and Steel Making

In this process, some selected flux such as limestone, coke, iron ore and sinter are feed into the blast furnace. Iron ore is formed giving a product that contain more than 90% iron and is called pig iron. A blast of heated air in form of gas, liquid or powder is introduced through the opening which burn most of the coke to produce the heat required by the process. Figure 1 showed the main processes in the iron and steel industry. At the far left are the inputs for the integrated process, the far right displays the outputs. Blast furnace gas is produced during the combustion of the coke in the blast furnaces (U.S. EPA, 2012). The coke is typically recovered and used as a fuel within the plant in other steel industrial processes. Coke can be transferred offsite and used as energy source in the furnace and the blast furnace gas is combusted to heat blast air. The final stage of refinement in composition of the iron end product is steel production. Processed iron ore from the blast furnace containing about 4% carbon and other impurities present need to be reduced for most application. The process involved addition of

different alloying element giving a finish product of desired properties. The BF is used for processing most of the nation's iron production and the EAF which is mostly used for scraped material (DOE, 2000). In BOF, atmospheric oxygen is blown through the molten metal burning off most of the carbon content and the heat of reaction supplies energy to maintain the required temperature. The EAF produces molten steel mainly by melting the steel scrap. However in some instances, direct reduced iron and pig iron are added to the charge (U.S. EPA, 1994; Rainer *et al.*, 2013 and U.S. EPA, 2012).

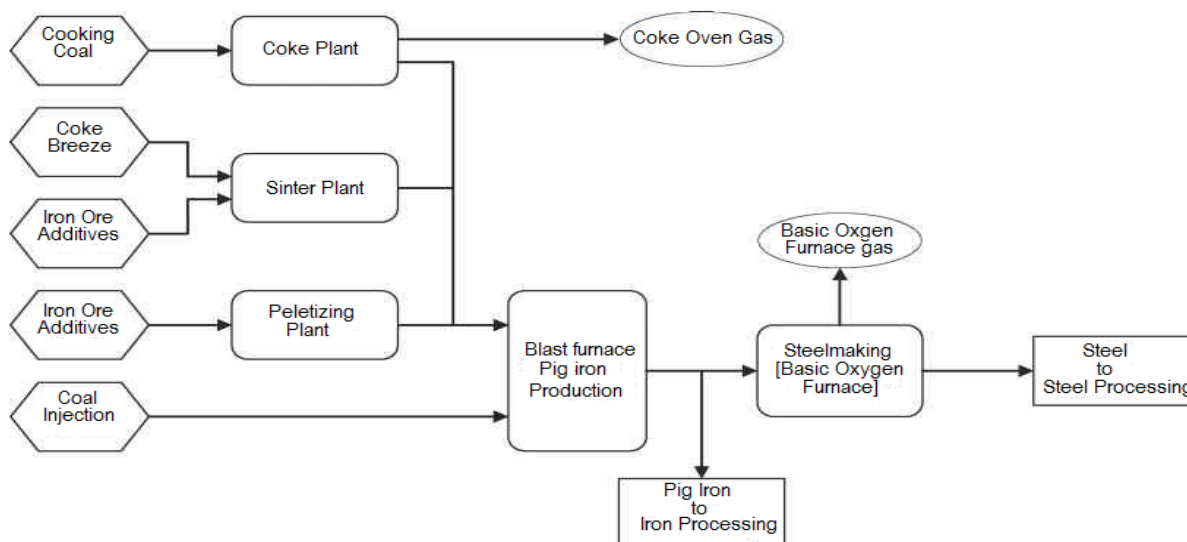
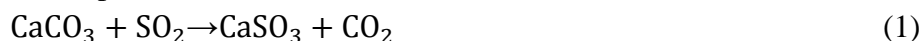


Figure 1: Main processes flow diagram for integrated iron and steel production (CPW, 2012)

2. Emissions from Iron and Steel Making

In iron and steel production, various pollutants are being emitted to the atmosphere. These pollutants are nitrogen oxides, sulphur oxides, carbon oxides ammonia, particulate matter, volatile organic compounds, slag, sludge, waste water contaminants, hazardous waste and solid waste resulting in significant regional and global environmental problems (Wu *et al.*, 2015). The main pollutants emitted in iron and steel production are oxides of nitrogen and sulphur oxides. Most of the oxides of sulphur such as sulphur dioxide and sulphur trioxide are scrubbed by the limestone input in the furnace. The emission of sulphur dioxide is gradually reduced by its reaction with the alkali forming compounds of sulphate and carbon dioxide as shown in equation 1. DOE (2000) and MPRO (2005) reported that the most important pollutant in the production of iron and steel from raw material is oxides of nitrogen. It is the most harmful emission that react with volatile organic compounds (VOCs) to depletes ozone.



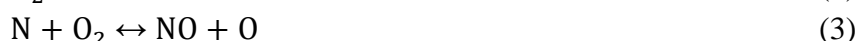
2.1 Mechanism involved in the Generation of NO_x

The processes that involved beneficiating iron ore and the recycling of iron bearing material such as sintering, pelletizing, coke making and steel finishing e.g. annealing, galvanizing and shaping requires high temperature application through the combustion of fossil fuel, are all sources of NO_x emissions (U.S. EPA, 1994). One significant source of NO_x comes from electricity generating units (EGUs) that often run heavily to meet demand for air conditioning on hot summer days in the U.S. EGUs also have smokestacks that emit at elevated heights and allow for long-range transport of pollutants (Vinciguerra *et al.*, 2017). NO_x is generated in iron making processes from crushing process, handling of iron ore and combustion of fuel in the furnace. The combustion of fuel requires

the mixture of air and the fuel. These react with oxygen to produce oxides of nitrogen NO_x as thermal NO_x, fuel NO_x and prompt NO_x. Fuel bound nitrogen and oxygen are contained in the coal and residual fuel. When these fuel are being combusted, NO_x is generated by all the three mechanism (U.S. EPA, 2008).

2.2 Generation of Thermal, Fuel and Prompt NO_x

Thermal NO_x is formed as a result of the thermal dissociation and subsequent reaction of nitrogen and oxygen molecules in the combustion air in the furnace. Most distillate oil and natural gas did not contain chemically bound fuel nitrogen. Essentially, all the NO_x produced in combustion of these fuel is thermal NO_x. At the combustion temperature the nitrogen and oxygen molecules are broken down into their respective atomic states. The Zeldovic reaction



shown in equation 2 and 3 indicated the formation of NO (U.S. EPA, 1999). The reaction depend on the decomposition of nitrogen and oxygen which increase exponentially with the temperature. The formation of NO_x is proportional to square root of concentration of oxygen, concentration of N₂ and residence time. Other factors affecting the formation of thermal NO_x includes velocity mixing, combustion air velocity and fuel injection angle (Toof, 1986).

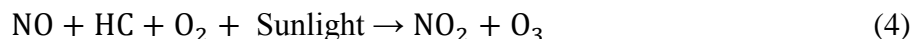
Fuel NO_x is generated by oxidation of nitrogen compounds contain in the fossil fuel which is combusted in the furnace producing molten iron. Fuel NO_x is formed below the temperature of thermal NO_x. Unlike Zeldovic model for thermal NO_x formation, the fuel bound nitrogen compounds react to form NO_x. Conversion rate of either solid phase or char reaction form NO_x depending on the air fuel ratio, flame temperature, char properties and gas phase reaction that results in cracking and revolution of volatile organic compounds (U.S. EPA, 1999).

Prompt NO_x mechanism involve the average formation of hydrogen cyanide (HCN), and then oxidize to NO (U.S. EPA, 1999 and MPRO, 2005). The formation of the prompt NO_x has a low temperature dependent and short lifespan of several micro seconds and its reaction occurs within the flame which is usually negligible when compared to the thermal NO_x. However, prompt NO_x level may become significant with ultra low NO_x burner.

2.3 Environmental Impact of NO_x

The reaction of oxides of nitrogen and VOCs in the presence of sunlight as shown in equation 4 emits directly into the air ground level "bad" ozone. NO_x reaction with oxygen and water forming nitric acid is deposited as acid precipitation affecting plants and animals on the land. This can be carried down to rivers thereby affecting the life of aquatic animals and other living things in the river (U.S. EPA, 2008). Acid and chemical deposition can occur as mist of rain, snow or fog, flows through the ground causing acidification and contamination of lakes and streams contribute to the damage of trees at high elevation and many sensitive forest soil. This deposit accelerates the decay of building materials, paints and irreplaceable buildings or statues and sculptures (U.S. EPA, 1999). Oxides of nitrogen undergoes reaction with elements forming nitric salts that leads to the formation of particulate matter which is inhalable and generally pass through human throat and nose causing serious damages to the lungs and heart. Other impacts include water quality deterioration, climate

changes, visibility impairment and toxic chemicals. Geng *et al* (2017) reported that air pollution is endangering the health of major cities in china through emission of NO_x, SO₂ and PM, reaching 20:44, 22:27 and 15 million of tons making china the largest emitter in the world.



3. Control Technologies of Emissions

The possible control technologies of oxides of nitrogen in iron and steel production involved techniques designed to reduce NO_x through combustion modification technologies such as low excess air (LEA), low NO_x burner and flue gas recirculation (FGR). Other techniques are designed to reduce the already generated NO_x from the waste effluent streams termed post combustion technologies (U.S EPA, 1994). This involved flue gas treatment techniques such as SCR and selective non catalytic reduction (SNCR) effectively reduce NO_x to molecular Nitrogen and water (ICAC, 1997a). Hybrid SCR/SNCR could be satisfactory compromise for NO_x reduction. Supplementary ammonia injection in the SCR section may be required for higher reductions (Srivastava *et al*, 2005). Other technologies for NO_x emissions control from reheat furnace, sintering furnace and coke ovens includes fuel switching and de-nitrification.

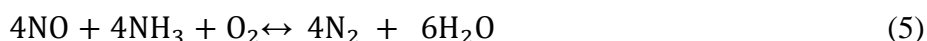
3.1 Combustion Modification Technologies

In this technology, air concentration is reduced to less than normal ratio required for the combustion processes in the furnace. This lower oxygen flame concentration thus reduce fuel and thermal NO_x generation while maintaining positive pressures in the furnace so as to prevent the influx of tramp air. The process is called low excess air. LEA reduces NO_x emissions up to 15% (U.S. EPA, 1994). LNB is designed to accommodate the changes in principle using LEA off staged combustion and FGR to the furnace for enhanced efficiency and reduction in emissions taking into account, any effect it may cause to the system. The objective of incorporating this system is to minimize NO_x generation while maintaining specific combustion level of the hydrocarbon content of the fuel at the furnace. FGR lowers flame temperature thus reducing emissions formed by thermal NO_x (U.S. EPA, 1999).

3.2 Post Combustion Technologies

3.2.1 Selective Catalytic Reduction (SCR) Technology

The SCR technology involves injection of ammonia into the flue gas containing oxides of nitrogen and convert the NO_x into molecular nitrogen (N₂) and water (H₂O) out of exhaust with improved performance. Temperature above 320⁰C is necessary to prevent ammonium salts from arising and blocking the catalytic converter (Rainer *et al.*, 2013). This method is effective at lower and wider temperature range of (260-485)⁰C depending on the type of catalyst used and the system design. The SCR technology is applicable to achieving NO_x reduction of up to 90% when properly implemented (Bradley, 2005; Srivastava *et al.*, 2005). In the SCR reactor, the flue gas temperature is controlled by mixing the flue gas from the economizer by pass with the flue gas exiting the economizer. The major reaction that occur in the SCR is shown in equation 5 and 6. The optimum temperature for the reaction occurs at 260-540⁰C. At higher temperature, the NH₄ oxidize to NO_x or NH₄NO₃ and NH₄NO₄ while for





lower temperature do not provide enough energy to start the reaction. Most of the catalysts used, are titanium oxides and vanadium oxides. However, ceramics, platinum, and zeolites are also used (U.S EPA, 1994). The SCR systems has been applied for NO_x control in coal fired power plants, other plants and refinery heaters, boilers and furnaces in chemical processing industries as well as municipal waste plant and incinerators (Cobb *et al.*, 1991).

3.2.2 Selective Non Catalytic Reduction (SNCR) Technology

SNCR involved the conversion of NO_x to molecular nitrogen and water by the injection of ammonia or urea into the flue gas stream while maintaining the proper range of temperature required about (930°C) and must be in proper ratio to the amount of NO_x present in the gas. This method could realise NO_x reduction of about 50% (Bradley, 2005). The injection of ammonia into the exhaust of a combustion system or the furnace above the combustion is commonly known as Exxon's Thermal De-NO_x process. This was first applied in oil and gas fired power plant (ICAC, 1997b). As no catalyst is used in the system, higher operating temperature is required between (982-1149°C) for urea and temperature of (871-982°C) for ammonia (ICAC, 1997b; Srivastava *et al.*, 2005). The overall chemical reaction for the urea and ammonia reducing nitric oxide to nitrogen and water is expressed in the equation 7 and 8 (Srivastava *et al.*, 2005; Mahmoudi *et al.*, 2010). Injection of Ammonia as well as urea can thermally be decomposed or oxidized at excessively high temperature greater than 1000°C. This will increase NO_x formation rather than destruction. At temperature below 1000°C, the ammonia slip becomes significant (ICAC, 1997b and IEA Clean Coal Center, 2011)



4. Results and Discussions

SCR utilizing ammonia or urea and catalyst is the most common NO_x mitigation technology usually applied in iron and steel industry due to its ability of reducing NO_x emission of up to 90% or more. The injection of catalyst in the technology fasten the rate of reaction as compared to SNCR, enough time at optimum temperature must be available for reducing the NO_x emissions. SCR plants installations; operations and capital cost are higher due to the catalyst involved. SNCR technology should spend much more on fuel required for residence time, good mixing and inquired climate change levy for emitting more pollutants as it is not so efficient in the control of NO_x emissions compared to SCR.

Because SCR is a dry technology there are no emissions to water. The waste generated which is deactivated catalyst can be reprocessed by the manufacturer (Rainer *et al.*, 2013). SCR system has been used frequently to reduce NO_x at higher level than SNCR. SCR is typically implemented on stationary source combustion unit requiring higher level of Nox reduction than achievable by SNCR or other combustion controls (U.S. EPA, 1994; U.S. EPA, 1999; MPRO, 2005 and U.S. EPA, 2015a). NO_x reduction at higher level is usually achieved by post combustion technology, combination of combustion modification with post combustion.

In this work, the data presented are based on limited data of existing reviews. Percentage reduction

and cost effectiveness of NO_x control technologies applied on coal, oil and gas fired plants for iron and steel production, process heating and other similar processes such as in

Table 1: Cost and percentage reduction of NO_x control technologies fired by coal

Nox Technology	Control Process/Type of Fuel	Percent Reduction	(\$/Year	Cost Effectiveness (\$/ton)	Reference
SCR	Coal fired	85	1990\$	8490	(U.S EPA, 1999)
SNCR	Coal fired	60	1994\$	3700	(U.S. EPA, 1994)
SCR+LNB	Coal fired	92	1990\$	10910	(U.S. EPA, 1999)
SNCR+LNB	Coal fired	80	1994\$	2900	(U.S. EPA, 1994)
LNB+FGR	Coal fired	60	1990\$	1790	(U.S. EPA, 1999)

Table 2: Cost and percentage reduction of NO_x control technologies fired by oil and gas

Nox Technology	Control Process/Type of Fuel	Percent Reduction	(\$/Year	Cost Effectiveness (\$/ton)	Reference
SCR	Oil/gas fired	85	1994\$	6900	(U.S. EPA., 2015b)
SNCR	Oil/gas fired	60	1990\$	3130	(U.S. EPA, 1999)
SCR+LNB	Oil/gas fired	94	2007\$	2910	(U.S. EPA, 2015a)
SNCR+LNB	Oil/gas fired	89	1990\$	3688	(MPRO, 2005)
LNB+FGR	Oil/gas fired	72	2004\$	4205	(MPRO, 2005)

Heating furnaces, boilers and power plants are shown in Table 2 and 3 respectively. It can be observed from Table 1 that 85% of NO_x is reduced by SCR system in 1990\$ with cost effectiveness of \$8490/ton. In 1994\$, the cost effectiveness of the SCR has drop to \$6900/ton with same 85% reductions achieved. In comparison with SNCR technology, \$3130/ton was achieved with 60% NO_x reduction in 1990\$ and \$3700/ton in 1994\$. In combined technology of combustion modification and post combustion, NO_x reduction of 92% for SCR and LNB systems has been attained with cost effectiveness of \$10910/ton of NO_x removed.

The percentage reduction of NO_x and cost effectiveness achieved by SCR and LNB is higher than the combined technology of SNCR and LNB which attained NO_x reduction of 80% with cost effectiveness of \$2900/ton as shown in Table 1. This is typically shown in Table 2 where higher percentage of NO_x reduction for SCR with LNB was achieved. Moreover, the cost effectiveness of combined technologies of SCR/LNB is lower compared to SNCR/LNB systems reported as \$2910/ton and \$3688/ton respectively (U.S. EPA, 2015a). It can be deduced that combined technology of SCR and LNB can reduce NO_x emission for up to 90% and above. The trend in continuous application of the SCR and SNCR systems shows a significant reduction in NO_x emission. However, their capital and control cost are usually high. Other impacts associated with the use of SCR and SNCR involved the release of some gases and acids to the systems which may require separate technology to reduce its emissions and the effects it will have on the systems.

5. Conclusion

The increase in the requirement for the reduction of NO_x emission from the combustion furnace of iron and steel mills requires a most effective approach for the development of the control technologies. SCR technology has a NO_x reduction of about 90%. This is because the SCR

performance can easily be controlled as it is specifically dependent on the type and amount of catalyst used in the system. Higher reaction efficiency of the SCR technology can be achieved by implementing the combination of combustion modification techniques with the SCR which can seriously reduce the material size and operating cost of the SCR systems. This is possible because the extent of reduction required by the SCR can be enhanced by the use of combustion modification technologies. Combine technology of SCR and low NO_x burner can highly reduce the emissions of NO_x over 90% with the minimum ammonia slip.

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